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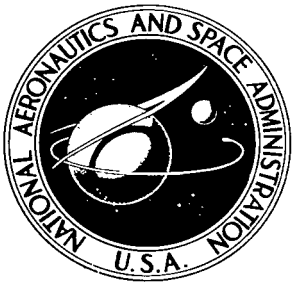

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ESTIMATION OF FISSION-PRODUCT GAS PRESSURE IN URANIUM DIOXIDE CERAMIC FUEL ELEMENTS

by Walter A. Paulson and Roy H. Springborn

Lewis Research Center
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ABSTRACT

Fission-product gas pressure in macroscopic voids was calculated over the temperature range of 1000 to 2500 K for clad uranium dioxide fuel elements operating in a fast neutron spectrum. The calculated fission-product yields for uranium-233 and uranium-235 used in the pressure calculations were based on experimental data compiled from various sources. The contributions of cesium, rubidium, and other condensible fission products are included with those of the gases xenon and krypton. At low temperatures, xenon and krypton are the major contributors to the total pressure. At high temperatures, however, cesium and rubidium can make a considerable contribution to the total pressure.

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SUMMARY

The fission-product gas pressure in macroscopic voids was calculated over the temperature range of 1000 to 2500 K for clad uranium dioxide ceramic fuel elements operating in a fast neutron spectrum. The contributions of the partial pressures of cesium, rubidium, and other fission products, which have generally been neglected in other investigations, are included in the pressure calculations. At low temperatures, xenon and krypton are the major contributors to the total pressure. However, it is shown that the partial pressures of cesium and rubidium at high temperatures can be as much as 70 percent of the partial pressures of xenon and krypton.

The total fission-product pressure is presented as a function of temperature for several values of a parameter θ. This parameter includes the fractional release of the fission products to the void, the number of fissions per cubic centimeter of fuel, and the ratio of void volume to fuel volume.

The calculation method can be applied to fission gas accumulating in a central void that is completely enclosed in the fuel and also to fuel with interconnected radial cracks that serve as the fission gas accumulator. It also can be applied to fuel elements with a void region between the fuel and cladding that serves as the fission gas accumulator and for the case of fuel vented to an external fission gas accumulator.

The calculated values of the fission-product yields, which were used in the pressure calculations, are presented in the appendix. These yields are from the fissioning of uranium-233 and uranium-235 by fission-spectrum neutrons and were determined on a per fission basis. They are based on some experimental data from various sources and were calculated on the basis of a reactor operating time of 50 000 hours. However, for pressure calculation purposes these yield fraction values are valid for operating times from 2000 hours to 100 000 hours, since they do not change appreciably.

INTRODUCTION

The choice of fast-spectrum nuclear reactors for possible space power application is a logical outcome of the requirement for compact power sources. Ceramic fuel (usually the oxide, carbide, or nitride form of a fissionable isotope) with a refractory-metal cladding is one type of fuel element that can be used in a fast reactor (ref. 1). The thickness of the fuel-element cladding is one of the design variables that affects reactor size. The fuel-element designer is faced with a weight penalty if the cladding is too thick or the possibility of cladding failure if it is too thin.

Buildup of fission-product gas pressure with exposure must be estimated if the fuel-cladding thickness is to be optimized. Underdesigned cladding can result in swelling and failure of the cladding caused by the fission-product gas pressure. The pressure level for a given fuel burnup depends on the amount of release of fission-product gases to void regions. This release, in turn, depends on the fuel, the burnup, the temperature level, and the temperature gradient from the fuel center to the fuel-cladding interface.

Uranium dioxide (UO_2) is one of the fuels considered for space power reactors. Uranium dioxide ceramic fuel elements have exhibited dimensional changes during irradiation (ref. 2). These dimensional changes can be the result of fuel swelling and of deformation of the cladding caused by internal fission-product gas pressure. This report presents a method for estimating the fission-product gas pressure in a clad uranium dioxide fuel element operating in a fast neutron spectrum. The contributions of the partial pressures of cesium, rubidium, and other fission products, which have generally been neglected in other investigations, are included in the estimation of the total pressure. Fuel swelling is not considered in this report.

Several models have been proposed as bases for calculating fission gas pressure and dimensional changes. Notley (ref. 3) developed a model to calculate the fission-product gas pressure in a UO_2 fuel element. Notley assumes that the fuel expands radially and axially, thereby causing interconnected cracks in the fuel. These cracks are assumed to provide the volume into which the fission gas is released. Fuel temperatures and crack volumes are calculated for an arbitrary number of radial increments. This technique is designed to approximate the actual temperature and crack-volume distribution in the fuel. The pressure-temperature (P-T) behavior of the fission gas released to the crack volume is treated in a relatively simple manner, however. In this model, an effective number of atoms (or moles) of fission gas released to the interconnected voids must be calculated. This calculation is based on the total number of fissions in the fuel and an estimated number of atoms released to the void per fission. The P-T behavior of this gas is then assumed to follow the ideal gas law. This ideal gas law treatment may not adequately approximate the actual P-T behavior over a wide temperature range because the number of atoms of gas released to the void per fission is temperature

dependent. As the temperature increases, the vapor pressure of the condensed fission products increases; hence, the number of atoms in the gas phase increases. At high temperature, it is shown herein that the vapor pressure of cesium and rubidium can make a significant contribution to the total pressure.

Duncombe, et al. (ref. 4), Friedrich (refs. 5 and 6), and Friedrich and Guilinger (ref. 7) developed a model that emphasizes the deformation of the fuel element. This model was incorporated into the CYGRO digital computer programs (refs. 5 to 7). One feature of the model included in CYGRO-2 is the calculation of the net force exerted by microscopic fission gas bubbles that are distributed throughout the fuel. The pressure in the bubbles is calculated by assuming that the fission gas can be represented as a single gas (i.e., xenon) which obeys van der Waals' equation of state. Diffusion coefficients are specified to allow for diffusion of the gas out of the bubbles. As in the case of Notley's model, a representative value of the number of atoms of fission gas released per fission must be estimated. The gas pressure in the macroscopic gaps, such as those between the fuel and the cladding, is not calculated but must be specified. Based on the net force, the model permits the fuel to densify or expand to eliminate gaps between the fuel and the cladding. The model also permits deformation of the cladding. Whereas the analysis in CYGRO-2 treats the microscopic gas bubbles in detail, this present investigation deals with fission gas pressure in macroscopic void regions. However, it can be concluded, based on arguments similar to those presented in the discussion of Notley's model, that the van der Waals' equation of state used in the CYGRO-2 computer program may not adequately approximate the P-T behavior in the microscopic gas bubbles.

In this report, the contribution of the partial pressures of cesium, rubidium, and other fission products not usually considered in pressure calculations are included along with xenon and krypton in calculating the fission-product gas pressure in macroscopic voids in clad uranium dioxide fuel elements.

The model used in calculating the fission-product gas pressure, the fission products considered in this investigation, the calculated pressure levels, and the fission-product yields for uranium-233 (U^{233}) and uranium-235 (U^{235}) are presented and discussed.

CALCULATIONAL MODEL

The method used for calculating the fission-product gas pressure can be applied to the following:

(1) A central void completely enclosed in the fuel

(2) Fuel with interconnected radial cracks

(3) A void region between the fuel and the cladding

(4) Fuel vented to an external fission gas tank

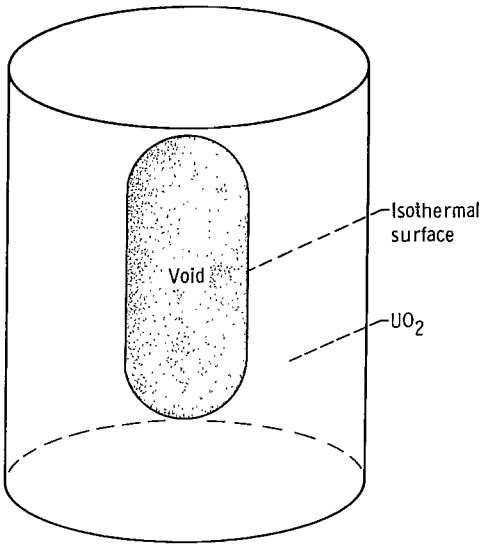


Figure 1. - Calculational model with central void.

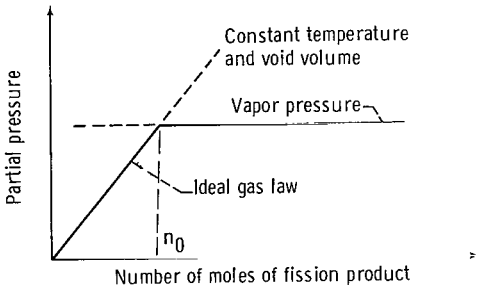


Figure 2. - Variation of partial pressure with amount of fission product produced.

In each of cases 1 to 3, it is understood that the void or the crack volume serves as the fission gas accumulator.

A conservative assumption from the standpoint of cladding design is that the fission products are at the temperature of an isothermal surface that defines a central void, as shown in figure 1. Each fission product that is not gaseous has an associated vapor pressure at the temperature of the fuel. If the fission yield of an element is too small, however, no condensed phase will be present, and the partial pressure exerted by the element will be less than its vapor pressure. In this case, the assumption was made that the fission product obeys the ideal gas law. This behavior is illustrated by figure 2. As the number of moles of fission product increases, the partial pressure increases, following the ideal gas law. When the number of moles exceeds n_0, a solid or liquid phase appears, and the partial pressure equals the vapor pressure at the temperature of the fuel.

Activity coefficients of the nongaseous fission products in uranium dioxide have not been determined; hence, the usual method of calculating the partial pressures of multicomponent mixtures is not available to the fuel-element designer. To simplify the analysis, the assumption was made that the liquid and solid fission products are completely immiscible. With this simplification, the partial pressure of each fission product is independent of its mole fraction.

FISSION PRODUCTS CONSIDERED IN CALCULATIONS

Chemical Combination of Fission Products

The fission-product yields for both U^{233} and U^{235} are shown in the appendix (tables V and VI). The fission-product yields used in this report are defined as the number of atoms of an element produced on a per fission basis after a reactor operating time of 50 000 hours. There is only a small difference between these yields and those at 2000 hours or at 100 000 hours; hence, the results of this investigation are applicable over this range of reactor operating times as discussed in more detail in the appendix.

Two atoms of oxygen become available for chemical combination for each fission. The oxygen combines with the more reactive fission products. Belle (ref. 8) tabulated the individual stable fission-product oxides based on an analysis of the Gibbs free energies of formation. The oxides are as follows: lanthanum oxide (La_2O_3), yttrium oxide (Y_2O_3), cerium oxide (Ce_2O_3), praseodymium oxide (Pr_2O_3), neodymium oxide (Nd_2O_3), promethium oxide (Pm_2O_3), europium oxide (Eu_2O_3), samarium oxide (Sm_2O_3), strontium oxide (SrO), barium oxide (BaO), zirconium dioxide (ZrO_2), stannic oxide (SnO_2), antimony trioxide (Sb_2O_3), and molybdenum dioxide (MoO_2). Because of the relatively low vapor pressure of these oxides, their contributions to the total pressure are neglected.

In addition to oxygen, some of the fission-product bromine and iodine may combine with cesium. For this analysis, however, the chemical combination of iodine and bromine is neglected.

Gaseous Fission Products

Xenon and krypton are the two fission products usually considered in fission-product pressure calculations because they are gaseous at room temperature and their yields are relatively large. Iodine and bromine are also gaseous at the operating temperature of UO_2 fuel elements considered in this report. In addition, cesium and rubidium are in

the gaseous phase in regions of the fuel that are above their critical temperatures. The critical temperature of cesium is about 2150 K and that of rubidium about 2170 K.

Solid and Liquid Fission Products

The remaining fission products considered herein are solid or liquid over the temperature range investigated. These elements are tellurium, selenium, cadmium, and also cesium and rubidium in regions that are below their critical temperatures. The partial pressure of solids and liquids is the vapor pressure at the temperature of the fuel, except as discussed in the section CALCULATIONAL MODEL.

The equation relating vapor pressure p and temperature T used in the pressure calculations was

$$\log p = \frac{A}{T} + B \log T + C$$

The constants A, B, and C for tellurium and selenium were determined using vapor pressure-temperature data found in the literature. These constants for cadmium were obtained from reference 9. In the case of cesium and rubidium, the data of Bonilla, et al. (ref. 10) were used. These data are valid to 100 psia (6.89×10^5 N/m^2 abs). The vapor pressures at high temperatures generally require an extrapolation over a wide range and may introduce moderate errors in the values of calculated pressure.

The values of the constants are as follows:

Element	A	B	C
Cs	-3703	0	6.822
Rb	-3880	0	6.927
Cd	-5819	-1.257	12.29
Te	-5680	0	7.39
Se	-5500	0	8.65

These values yield pressure in millimeters of mercury when the temperature is Kelvin.

Correction for Dimer in Cesium Vapor

In the regions where cesium has been assumed to obey the ideal gas law, a correction has been included to account for the fraction of the cesium that is in dimer form

$$p_{Cs} = \frac{n_{Cs}RT}{V} F(T)$$

where p_{Cs} is the partial pressure of cesium, n_{Cs} is the number of moles of cesium, R is the gas constant, T is the absolute temperature, V is the volume occupied by the cesium, and F(T) is the temperature-dependent correction factor for the cesium dimer.

The constants in the expression for F(T)

$$F(T) = 10^{(-0.1559 \log_{10} T + 0.433)}$$

were determined for the temperature range of interest from the data in reference 11. This correction yields a lower partial pressure than would be calculated if 100 percent monomer were assumed.

Cesium and Rubidium Distribution

The fractional release of the fission products to the interconnected void is one of the variables that affects the total pressure level. Notley (ref. 3), Duncombe (ref. 4), and the present investigation require an estimate of this release fraction. The fractional release of xenon and krypton can be determined from postirradiation measurements. The fractional release of cesium and rubidium, the other major contributors to the total fission-product gas pressure, must also be determined. Because cesium and rubidium are both in a condensed phase at the temperatures that exist during postirradiation measurements, their effective release to the void may not be as easily determined as that of xenon and krypton.

The yield of cesium and rubidium is about the same as that of xenon and krypton (see table VI in the appendix). Almost all the cesium and rubidium are decay products of gaseous fission products. The precursors and daughter products are as follows:

Parent	Half-life	Daughter
Xe^{133}	5.27 days	Cs^{133}
Xe^{135}	9.2 hr	Cs^{135}
Xe^{137}	3.9 min	Cs^{137}
Kr^{85}	4.4 hr (81 percent)	Rb^{85}
Kr^{85}	10.6 yr (19 percent)	Rb^{85}
Kr^{87}	78 min	Rb^{87}

The precursors of both cesium and rubidium migrate up the temperature gradient as xenon and krypton. Hence, the proportion of cesium and rubidium released could be expected to be similar to that of xenon and krypton. The migration rate depends on the temperature level and the temperature gradient. This rate can be calculated by using a method described by Nichols (ref. 12). Morgan, et al. (ref. 13) report measured values of the radial distribution of fission-product xenon and cesium in UO_2 fuel elements. The depletion of both these fission products in the region of grain growth was similar. Therefore, for the conditions of the irradiation described by Morgan, the fractional release of the four major contributors to the fission-product gas pressure can be assumed to be the same in the region of grain growth. This assumption may not be valid in other regions in the fuel or at other operating conditions. Because of the lack of additional experimental data, however, it was assumed in the present analysis that the fractional release is the same for all fission products considered.

Morgan also found that eventually the cesium migrated to the colder regions of the fuel. This migration may have occurred through interconnected cracks in the fuel. Based on Morgan's investigation, it can be expected that cesium (as well as other condensibles) may migrate from one void region to a cooler, interconnected void region where it can condense if the temperature is below the critical temperature of the element.

CALCULATED PRESSURE IN UO_2 CERAMIC FUEL

The pressure level depends on the total fissions, the fission yield of each element, the void volume, the fractional release of the fission products from the fuel matrix to the void, and the temperature level. The yields used in this calculation are based on the assumption of a fast spectrum and are shown in the appendix (table VI).

The total fission-product pressures calculated using the central void model of figure 1 are shown in figure 3 and are plotted against temperature for various values of the parameter θ. This parameter includes the fractional release of fission products, fissions per cubic centimeter of fuel, and the ratio of void volume to fuel volume.

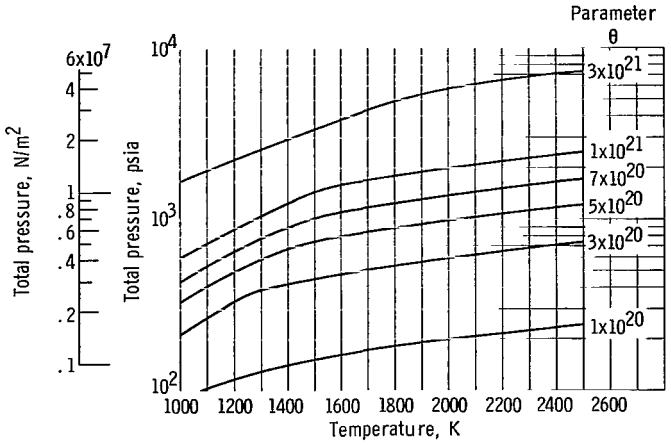


Figure 3. - Total fission-product gas pressure as function of temperature.

$\theta = \frac{[\text{Fissions per cm}^3 \text{ of fuel}][\text{Fractional release}]}{\text{Void volume/Fuel volume}}$.

$$\theta = \frac{[\text{Fissions per cm}^3 \text{ of fuel}][\text{Fractional release}]}{\frac{\text{Void volume}}{\text{Fuel volume}}}$$

The partial pressure for elements above their critical temperature was calculated by using the ideal-gas law:

$$p_i = \frac{n_i RT}{V} = \frac{\theta X_i RT}{N_A}$$

where p_i is the partial pressure of element i, n_i is the number of moles of element i, R is the gas constant, T is the absolute temperature, V is the void volume, N_A is Avogadro's number, X_i is the yield per fission for element i and is taken from table VI in the appendix, and θ is defined above.

In the case of fission products below their critical temperature, the pressure was calculated by using both the ideal gas law and the vapor pressure relation:

$$p_i = 10^{(A/T + B \log_{10} T + C)}$$

where the constants A, B, and C are discussed in the section Solid and Liquid Fission

Products. The partial pressure for the elements below their critical temperatures was then taken as the lesser of the two pressures calculated by using the ideal gas law or the vapor pressure relation. The total pressure in the void volume is merely the sum of the partial pressures of the fission products considered.

Generally, the designer will know the fuel burnup. The ratio of the effective void volume to fuel volume can be calculated from the as-fabricated fuel element and the relative thermal expansion of fuel and cladding. A conservative estimate for the purpose of investigating the fission gas pressure effect on cladding design is to assume the fractional release is 1.0. A fractional release of less than 1.0 may result in fuel swelling that can cause deformation of the cladding. However, fuel swelling effects are not considered in this report.

Relative Effect of Individual Fission Products

Figure 4 shows the individual contributions of the fission products to the total pressure based on the model shown in figure 1. At 2500 K, the partial pressure of cesium and rubidium is as much as 70 percent of the partial pressure of xenon and krypton.

Effects of Radial Cracks

The model used to calculate the pressure levels shown in figure 3 assumes the fission products are at the temperature of an isothermal surface that defines a central void and ignores any radial cracks. The effect of cracks would be to permit the cesium

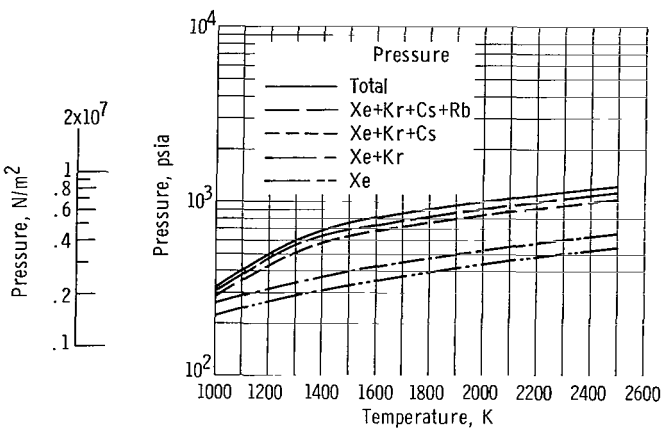


Figure 4. - Contribution of individual fission products of U^{235} to total pressure. $\theta = 5x10^{20}$.

and rubidium to migrate to regions of the fuel near the cladding and to condense at the lower temperatures (assuming the cladding is at a temperature below the critical temperatures of cesium and rubidium). This would reduce the total pressure level. The pressure levels for a model that incorporates radial cracking can be obtained by correcting figure 3 for the decreased cesium and rubidium partial pressures. This correction can be easily made by using figures 5 and 6, which show the partial pressures of cesium and rubidium as a function of temperature and the parameter θ.

Vented Fuel Element

The total pressure levels shown in figure 3 are also applicable to UO_2 ceramic fuel elements vented to an external accumulator. In this case, the temperature to be used with figure 3 is the temperature of the accumulator walls.

Void Between Fuel and Cladding

For the case of a void between the fuel and the cladding, figure 3 can be used directly. In this case, the temperature to be used is an average of the fuel surface and cladding temperatures.

SUMMARY OF RESULTS

Fission-product gas pressure was calculated over the temperature range of 1000 to 2500 K for clad uranium dioxide fuel elements operating in a fast neutron spectrum. Fission-product pressure as a function of temperature for several values of a parameter θ are presented. This parameter includes the fractional release of the fission products to the void, the number of fissions per cubic centimeter of fuel, and the ratio of void volume to fuel volume.

At low temperatures, xenon and krypton are the major contributors to the total pressure. At high temperatures, however, the partial pressures of cesium and rubidium can be as much as 70 percent of the partial pressures of xenon and krypton.

The calculation method can be applied to fission gas accumulating in a central void that is completely enclosed in the fuel and also to fuel with interconnected radial cracks that serve as the fission gas accumulator. It also can be applied to fuel elements with a void region between the fuel and the cladding that serves as the fission gas accumulator and for the case of fuel vented to an external fission gas accumulator.

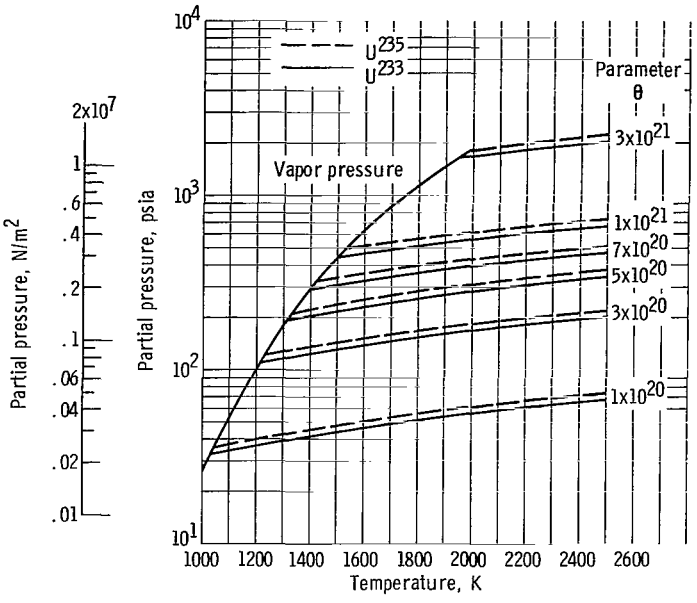


Figure 5. - Cesium partial pressure as function of temperature for U^{233} and U^{235}.

$$\theta = \frac{\text{[Fissions per cm}^3\text{ of fuel][Fractional release]}}{\text{Void volume/Fuel volume}}$$

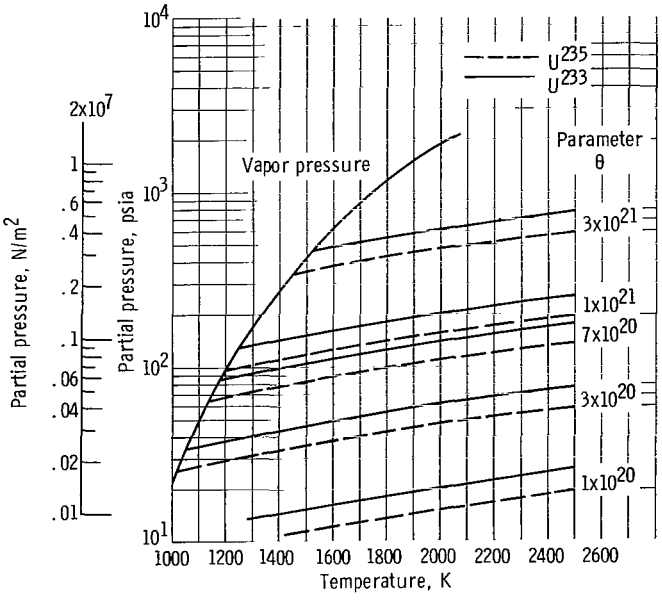


Figure 6. - Rubidium partial pressure as function of temperature for U^{233} and U^{235}.

$$\theta = \frac{\text{[Fissions per cm}^3\text{ of fuel][Fractional release]}}{\text{Void volume/Fuel volume}}$$

The calculated fission-product yields for uranium-233 and uranium-235 used in the pressure calculations were based on experimental data from various sources. They are based on a reactor operating time of 50 000 hours but are also valid over the range of 2000 to 100 000 hours.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, July 2, 1968,
120-27-06-17-22.

APPENDIX - FISSION-PRODUCT YIELDS FROM FISSIONING OF URANIUM-233 AND URANIUM-235 INDUCED BY FISSION-SPECTRUM NEUTRONS

To calculate the pressure due to the fission products it is necessary to know the fission-product yields. A fission-product yield, in this sense, is the number of atoms of an element produced on a per fission basis for the reactor operating time of interest.

The reactor operating time must be taken into consideration, since the initial fission products are radioactive and decay into other isotopes. The amount of radioactive parent remaining and the amount of daughter product formed depend on the length of reactor operation.

Fission-product yields were determined for U^{233} and U^{235}, since both these fuels are of interest for the fast-spectrum nuclear reactor being considered for space power applications (ref. 14).

The fission-product yields were determined in the following steps:

(1) For U^{233}, mass-number yields were obtained by Bunney and Scadden (ref. 15) from their own experiments and from those of Bonyushkan, et al. (ref. 16).

(2) They then "reflected" these mass-number yields to obtain additional values.

(3) A curve of yield against mass number was then constructed by Bunney and Scadden, using the original data and the reflected points.

(4) For U^{235}, the authors obtained mass-number yield data from reference 17, which is a listing of data from various sources. The points were then reflected and a curve drawn, following the technique employed by Bunney and Scadden.

(5) For both U^{233} and U^{235}, a complete listing was made of mass-number yields for each mass number from 72 to 161, using the curves that were developed.

(6) The isotopic yield fractions in each mass-number chain were then determined, on the basis of a reactor operating time of 50 000 hours.

(7) Finally, the individual isotopic yields were grouped by elements and added together. These were the yields considered for the fission gas pressure calculations.

Uranium-233 Experimental Fission-Product Mass-Number Yields

A mass-number yield is the sum of the individual isotopic fission-product yields of a given mass number. In the following illustration of the mass chain for mass-number 147, the mass-number yield is the sum of the yields of barium (Ba), lanthanum (La), Ce,

praeseodymium (Pr), neodymium (Nd), promethium (Pm), and samarium (Sm)[1], at any given reactor operating time.

$$_{56}Ba^{147}\ (1.0\ \text{sec}) \longrightarrow {}_{57}La^{147}\ (2.0\ \text{sec}) \longrightarrow {}_{58}Ce^{147}\ (1.2\ \text{min}) \longrightarrow$$

$$_{59}Pr^{147}\ (12.0\ \text{min}) \longrightarrow {}_{60}Nd^{147}\ (11.1\ \text{day}) \longrightarrow {}_{61}Pm^{147}\ (2.6\ \text{year}) \longrightarrow$$

$$_{62}Sm^{147}(\text{stable})$$

The method of decay in this example and for all other fission products was considered to be by loss of a negative beta particle. As can be seen from this illustration, the mass number remains constant for each decay step in the mass chain. The mass-number yields, therefore, do not change during the decay process. Thus, it is convenient to consider fission-product yields by mass numbers.

Bunney and Scadden obtained their experimental mass-number yields by determining the yield of the last (in most cases) radioactive isotope of the mass chain. For mass-number 147 this was Nd. They used mass-number yields determined experimentally by Bonyushkan, et al. (ref. 16) to supplement their own experimentally determined values. The experimental yields from both sources are listed in table I.

Uranium-233 Reflected Mass Numbers

Bunney and Scadden calculated additional mass-number yields by assuming that the plot of yield against mass number is exactly symmetrical; that is, for each "heavy" mass-number yield there is an identical "light" mass-number yield and vice versa. These calculated mass numbers are called "reflected" mass numbers since the plot is symmetrical about some mass number (115.75 for U^{233}, and 116.75 for U^{235}).

The reflected mass number was calculated by considering that for each fission, two fission products are formed plus an average of 2.5 neutrons. The mass number of the reflected fission product was then found from the following expression for the average mass-number balance for the fission process:

---

[1]As indicated in ref. 18, negligible amounts of the longer half-life isotopes are formed directly as primary fission products. For mass-number 147 these are Nd, Pm, and Sm.

TABLE I. - FISSION-PRODUCT MASS-NUMBER YIELD DATA AND REFLECTED YIELDS FROM FISSIONING OF URANIUM-233 INDUCED BY FISSION-SPECTRUM NEUTRONS

Mass number	Experimental data from reference 15	Reflected yields from reference 15	Experimental data from reference 16	Reflected yields from reference 16	Mass number	Experimental data from reference 15	Reflected yields from reference 15	Experimental data from reference 16	Reflected yields from reference 16
	Fractional yield, [a] atoms/fission					Fractional yield, [a] atoms/fission			
$70\frac{1}{2}$	---	5×10^{-6}	---	---	$116\frac{1}{2}$	---	1.01×10^{-3}	---	5.6×10^{-4}
$72\frac{1}{2}$	---	1.8×10^{-5}	---	---	$120\frac{1}{2}$	---	1.3	---	8.4
$74\frac{1}{2}$	---	1.05×10^{-4}	---	---	$125\frac{1}{2}$	---	---	---	1.6×10^{-3}
$75\frac{1}{2}$	---	1.82	---	---	$128\frac{1}{2}$	---	---	---	4.13
$78\frac{1}{2}$	---	1.26×10^{-3}	---	---	129	---	---	1.57×10^{-2}	---
$80\frac{1}{2}$	---	3.48	---	---	132	5.17×10^{-2}	---	4.36	---
$82\frac{1}{2}$	---	8.22	---	---	$132\frac{1}{2}$	---	---	---	4.75×10^{-2}
$84\frac{1}{2}$	---	1.72×10^{-2}	---	---	137	---	---	6.28×10^{-2}	---
$87\frac{1}{2}$	---	4.19	---	---	140	---	---	6.31	---
$88\frac{1}{2}$	---	5.01	---	---	$140\frac{1}{2}$	---	6.61×10^{-2}	---	---
89	5.98×10^{-2}	---	6.30×10^{-2}	---	141	6.83×10^{-2}	---	6.74×10^{-2}	---
$90\frac{1}{2}$	---	6.83×10^{-2}	---	6.74×10^{-2}	$142\frac{1}{2}$	---	5.98×10^{-2}	---	6.30×10^{-2}
91	6.61×10^{-2}	---	---	---	143	5.01×10^{-2}	---	---	---
$91\frac{1}{2}$	---	---	---	6.31×10^{-2}	144	4.19	---	---	---
$94\frac{1}{2}$	---	---	---	6.28	147	1.72	---	---	---
99	---	---	4.75×10^{-2}	---	149	8.22×10^{-3}	---	---	---
$99\frac{1}{2}$	---	5.17×10^{-2}	---	4.36×10^{-2}	151	3.48	---	---	---
$102\frac{1}{2}$	---	---	---	1.57	153	1.26	---	---	---
103	---	---	4.13×10^{-3}	---	156	1.82×10^{-4}	---	---	---
106	---	---	1.6	---	157	1.05	---	---	---
111	1.3×10^{-3}	---	8.4×10^{-4}	---	159	1.8×10^{-5}	---	---	---
115	1.01	---	5.6	---	161	5×10^{-6}	---	---	---

[a]Fractional mass numbers for reflected yields result from mass-number balance equation, for which 2.5 was used for average number of neutrons emitted per fission.

Mass number of fuel + 1 Neutron = Mass number of measured fission product

+ Mass number of reflected fission product + 2.5 Neutrons

The reflected mass numbers for U^{233} are listed in table I.

Although only whole neutrons can be emitted during fission, the same number of neutrons is not always emitted. A statistical average of the number of neutrons emitted per fission can therefore be a fractional number. Since this average is approximately 2.5, the reflected mass numbers computed from the preceding equation are also fractional numbers. Although a fractional mass number does not exist physically, it does have a valid statistical meaning.

Uranium-233 Mass-Number Yield Curve

Using the experimentally determined mass-number yields and the reflected points, Bunney and Scadden constructed a curve of fission yield as a function of mass number for U^{233}. The fission yield is plotted on a logarithmic scale since the fractional yields range from about 10^{-5} to more than 0.06 atoms per fission. The curve is shown in figure 7.

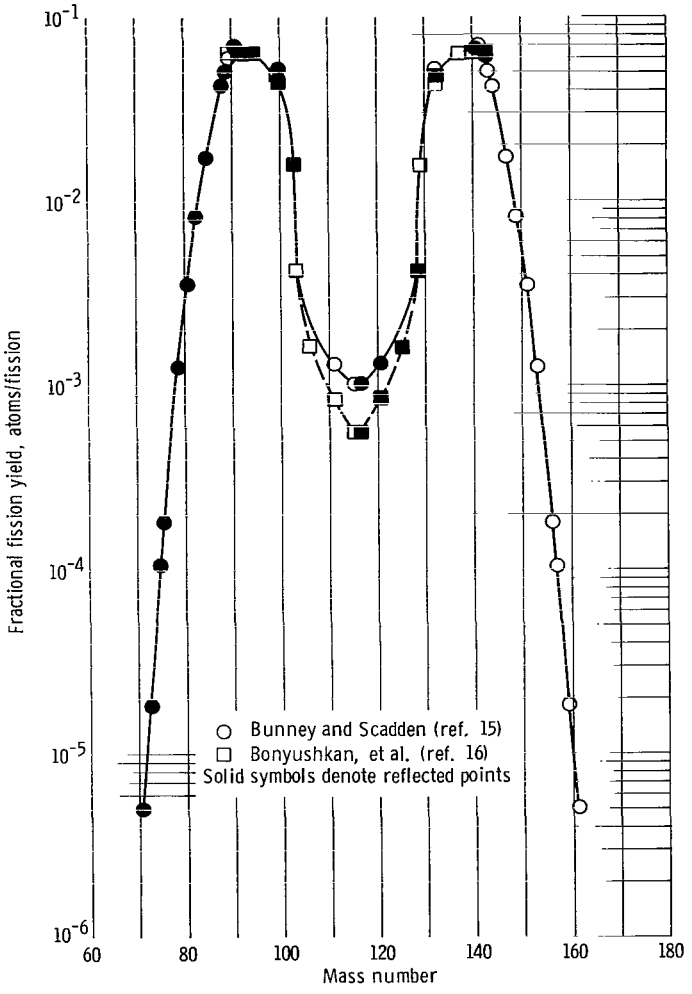


Figure 7. - Mass yields of fission-spectrum neutron fission of U^{233}.

Uranium-235 Fission-Product Mass-Number Yield Curve

Following the technique employed by Bunney and Scadden, the authors developed the mass-number yield curve for U^{235}. The experimental yield values for U^{235} are an average of data obtained from various sources and assembled by Zysin, Lbov, and Sel'chenkow (ref. 17). The mass numbers were reflected to obtain additional yields, and the mass-number yield curve was constructed. The averaged yield values from the experimental data and the calculated reflected mass numbers are listed in table II. The mass-number yield curve is shown as figure 8.

TABLE II. - FISSION-PRODUCT MASS-NUMBER YIELD DATA AND REFLECTED YIELDS FROM FISSIONING OF URANIUM-235 INDUCED BY FISSION-SPECTRUM NEUTRONS

Mass number	Fractional yield data,[a] atoms/fission	Reflected mass number	Mass number	Fractional yield data,[a] atoms/fission	Reflected mass number
89	5.0×10^{-2}	$144\frac{1}{2}$	129	[b] 5.5×10^{-3}	$104\frac{1}{2}$
90	5.0	$143\frac{1}{2}$	132	5.35×10^{-2}	$101\frac{1}{2}$
95	6.78	$138\frac{1}{2}$	137	6.59	$96\frac{1}{2}$
97	6.55	$136\frac{1}{2}$	140	5.5	$93\frac{1}{2}$
99	6.13	$134\frac{1}{2}$	141	6.1	$92\frac{1}{2}$
103	3.48	$130\frac{1}{2}$	144	5.13	$89\frac{1}{2}$
105	1.45	$128\frac{1}{2}$	147	2.4	$86\frac{1}{2}$
106	9.50×10^{-3}	$127\frac{1}{2}$	149	1.23	$84\frac{1}{2}$
109	1.46	$124\frac{1}{2}$	153	1.9×10^{-3}	$80\frac{1}{2}$
111	4.6×10^{-4}	$122\frac{1}{2}$	156	2.4×10^{-4}	$77\frac{1}{2}$
112	4.1	$121\frac{1}{2}$	159	3.4×10^{-5}	$74\frac{1}{2}$
115	3.3	$118\frac{1}{2}$	161	4.6×10^{-6}	$72\frac{1}{2}$

[a] Fractional yield value is average of data listed by Zysin, et al. (ref. 17).

[b] Data point for mass-number 129 is omitted in fig. 8 since it is only a partial yield for the mass number.

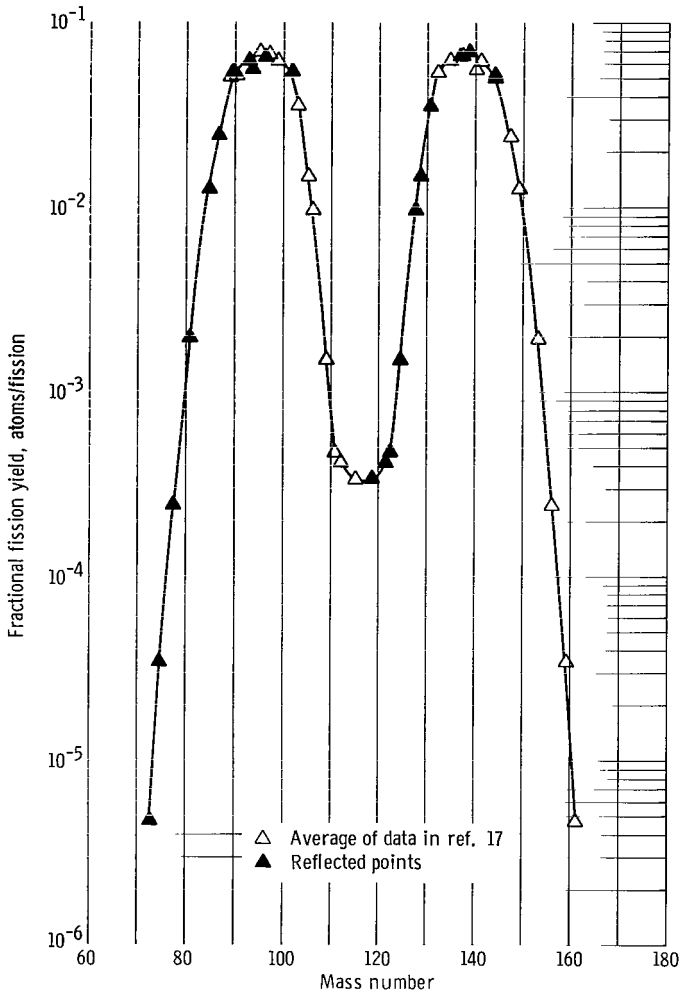


Figure 8. - Mass yields of fission-spectrum neutron fission of U^{235}.

Uranium-233 and Uranium-235 Mass-Number Yields For Each Mass Number From 72 to 161

From the curves that were constructed, all the mass-number yields Z were obtained for mass numbers from 72 to 161, as listed in table III in units of atoms per fission. It was assumed that all mass numbers were present in the fission products, just as Kochendorfer assumed this for thermal neutron fission of U^{235} (ref. 18). The resultant total yields were about 1.92 for U^{233} and 2.09 for U^{235}, close to the 2.00 atoms expected from the binary fission of a heavy nucleus.

TABLE III. - FISSION-PRODUCT MASS-NUMBER YIELDS AND ISOTOPES FROM FISSIONING OF URANIUM-233 AND URANIUM-235 INDUCED BY FISSION-SPECTRUM NEUTRONS

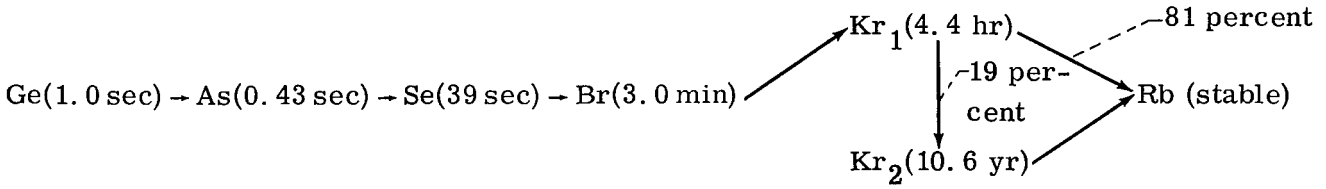
Mass number	Fractional mass number yields, Z, atoms/fission		Final stable isotope, or radioactive parent and daughter
	U^{233}	U^{235}	
72	1.4×10^{-5}	---------	Ge
73	3	7×10^{-6}	Ge
74	6	2×10^{-5}	Ge
75	1.2×10^{-4}	4.5	As
76	2.5	9	Ge
77	5	1.8×10^{-4}	Se
78	1.0×10^{-3}	3.5	Se
79	1.6	6	Se
80	2.6	1.2×10^{-3}	Se
81	4.4	2.4	Br
82	6.8	4.1	Se
83	1.00×10^{-2}	6.8	Kr
84	1.50	1.05×10^{-2}	Kr
85	1.95	1.50	Rb(81 percent) + Kr(19 percent)[a]
Z_1	1.58	1.21	Rb
Z_2	3.7×10^{-3}	2.9×10^{-3}	Kr(10.6 years) → Rb
86	2.65×10^{-2}	2.05×10^{-2}	Kr
87	3.50	2.70	Rb
88	4.80	3.50	Sr
89	6.14	4.50	Sr(50 days) → Y
90	6.40	5.20	Sr
91	6.61	5.60	Y(58 days) → Zr
92	6.60	6.00	Zr
93	6.50	6.30	Zr
94	6.40	6.50	Zr
95	6.05	6.78	Zr(65 days) → Nb(35 days) → Mo
96	5.85	6.60	Zr
97	5.50	6.55	Mo
98	5.10	6.30	Mo
99	4.75	6.13	Tc
100	4.20	5.75	Mo
101	3.25	5.50	Ru
102	2.40	4.70	Ru
103	1.30	3.48	Ru(40 days) → Rh
104	3.3×10^{-3}	2.30	Ru
105	2.6	1.45	Pd
106	2.15	9.5×10^{-3}	Ru(365 days) → Pd
107	1.90	5.0	Pd
108	1.68	2.7	Pd
109	1.54	1.5	Ag
110	1.39	8×10^{-4}	Pd
111	1.30	5	Cd
112	1.20	4	Cd
113	1.13	4	Cd
114	1.05	4	Cd
115	1.01	3	In
116	1.01	3	Cd
117	1.05	3	Sn
118	1.12	3	Sn
119	1.17	3	Sn
120	1.26×10^{-3}	4×10^{-4}	Sn
121	1.36	4	Sn(50 percent) + Sb(50 percent)
Z_1	6.8×10^{-4}	2	Sn
Z_2	6.8	2	Sb
122	1.48×10^{-3}	4	Sn
123	1.65	7	Sb
124	1.83	1.2×10^{-3}	Sn
125	2.05	2.1	Sb(985 days) → Te
126	2.4	4.0	Sn(86.2 percent) + Te(13.8 percent)
Z_1	2.07	3.4	Sn
Z_2	3.3×10^{-4}	6×10^{-4}	Te
127	2.9×10^{-3}	8.0×10^{-3}	Te(105 days) → I
128	3.7	1.30×10^{-2}	Te
129	1.57×10^{-2}	1.90	I
130	2.80	3.00	Te
131	3.90	4.30	Xe
132	4.60	5.35	Xe
133	5.00	5.60	Cs
134	5.30	6.00	Xe
135	5.60	6.20	Cs
136	6.00	6.30	Xe
137	6.28	6.59	Cs
138	6.50	6.70	Ba
139	6.60	6.70	La
140	6.60	6.40	Ce
141	6.60	6.10	Ce(33 days) → Pr
142	6.40	5.80	Ce
143	5.40	5.50	Nd
144	4.19	5.13	Ce(285 days) → Nd
145	3.10	4.00	Nd
146	2.30	3.10	Nd
147	1.72	2.40	Pm(950 days) → Sm
148	1.18	1.80	Nd
149	8.2×10^{-3}	1.23	Sm
150	5.4	8.2×10^{-3}	Nd(50 percent) + Sm(50 percent)
Z_1	2.7	4.1	Nd
Z_2	2.7	4.1	Sm
151	3.5	5.4	Sm
152	2.2	3.3	Sm
153	1.26	1.9	Eu
154	7.0×10^{-4}	1.0	Sm
155	3.5	5×10^{-4}	Eu(622 days) → Gd
156	1.8	2.4	Gd
157	1.1	1.3	Gd
158	4×10^{-5}	6×10^{-5}	Gd
159	2	3.4	Tb
160	1	1.3	Gd
161	5×10^{-6}	4.6×10^{-6}	Dy
Total	1.924	2.095	

[a]Some decay schemes have branching isotopes. Fraction of yield for each branch is noted in parentheses.

Isotopic Yield Fractions in Each Mass Chain

For the pressure calculations, it is necessary to determine the amount of each fission-product isotope present at the end of the reactor operating period. The isotopes are then added together by elements to determine their contribution to pressure. The number of atoms of a fission-product isotope present per fission is the isotope yield fraction and depends primarily on the fractional yield of its mass number and the characteristics of the decay scheme.

The following decay scheme for mass-number 85 shows the isotopes and half-lives in the chain and also illustrates a decay scheme that has branching fractions (Kr_1 branching to Kr_2 and Rb). This decay scheme and the other decay schemes used in the calculations were taken to be the same as that for thermal neutron fission of U^{235}, as presented by Kochendorfer (ref. 18).



As in this example, the first few fission products in the mass chain are typically highly radioactive and decay quickly into more stable isotopes by negative beta decay. These highly radioactive isotopes do not make significant contributions to the yields of their element families for long-term reactor operation.

Isotopes which do contribute to the yield of an element are either stable isotopes or radioactive isotopes which exist long enough to make their influence felt. For a reactor operating time of 50 000 hours, these isotopes were calculated to be those with half-lives greater than about 16 days.

For a mass chain in which all precursors of a stable isotope had half-lives less than 16 days, the entire mass chain yield is essentially the yield for that stable isotope. (A "stable" isotope for this analysis was calculated to be one with a half-life greater than 28 years.)

For isotopes with half-lives between 16 days and 28 years, the yield fractions of the radioactive parent and the daughter had to be calculated, since they both contribute to the total yield. And they had to be calculated for a specific reactor operating time, because the yield fraction of the parent (on a per fission basis) decreases with time and that of the daughter increases with time. The reactor operating time for these calculations was chosen as 50 000 hours. In the preceding mass chain, the fractional yield of the radioactive isotope krypton (Kr_2) and the daughter rubidium (Rb) had to be calculated.

The fractional isotopic yields of a radioactive parent and daughter depend on (1) the rate of formation of the parent by the fission process, (2) the rate of radioactive decay of the parent, (3) the rate of transmutation of a fission product into a different isotope by neutron capture (in general), and (4) the reactor operating time.

If the radioactive parent isotope is designated A, the rate of formation of A is

$$\frac{dA}{dt} = Z_A \Sigma_f \varphi$$

where

Z_A atoms of isotope A produced per fission (After equilibrium is established, it is equal to the yield of its mass number for decay chains in which no branching occurs. When branching occurs, as it does in the example for mass-number 85, the mass-number yield is divided into branching fractions Z_1 and Z_2.)

$\Sigma_f \varphi$ rate of fissioning of fuel per unit volume, fissions/(cm^3)(sec)

The rate of radioactive decay of the parent A is

$$\frac{dA}{dt} = -\lambda_A A$$

where

λ_A radioactive decay constant of isotope A

A amount of isotope A present at any time

In general, fission products can also be removed from the system by transmutation into a different isotope by neutron capture. The rate of removal is

$$\frac{dA}{dt} = -\sigma_A \varphi A$$

where

σ_A microscopic absorption cross section for isotope A

φ neutron flux

In the fast-spectrum nuclear reactor, however, this loss is small[2] compared with the rate of formation and the rate of radioactive decay and was therefore neglected in the calculations.

Combining the expressions for the formation rate and the radioactive decay rate results in the net rate of change in the amount of isotope A per unit volume with time t in the reactor given by

$$\frac{dA}{dt} = Z_A \Sigma_f \varphi - \lambda_A A$$

With the initial condition that no fission products exist at reactor startup, this equation can be solved for A(t), the amount of isotope A present at any reactor operating time t.

$$A(t) = \frac{Z_A \Sigma_f \varphi}{\lambda_A} \left(1 - e^{-\lambda_A t}\right)$$

This equation can now be divided by the total number of fissions occurring during the reactor operation $\Sigma_f \varphi t$ to obtain the yield on a per fission basis. For any isotope, then,

$$Y = Z\left(\frac{1 - e^{-\lambda t}}{\lambda t}\right)$$

where Y is the isotopic yield fraction after a reactor operating time t (atoms of isotope/fission).

The yield fraction of the daughter isotope into which the parent decays is the difference between the fractional number of atoms of the parent produced per fission Z and the isotopic yield fraction Y of the parent after a reactor operating time t.

Table III presents the following results:

(1) Mass-number yields

(2) Division of mass-number yields for the branching fractions

(3) The final stable isotope for a mass number

(4) The radioactive parent and daughter products for isotopes with half-lives between 16 days and 28 years

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[2]The microscopic cross section σ of the fission products for 1-MeV neutrons is only about 0.045 barn (ref. 19).

TABLE IV. - FISSION-PRODUCT YIELDS OF RADIOACTIVE PARENTS AND DAUGHTERS AFTER 50 000 HOURS REACTOR OPERATION

Mass number	Parent radioactive isotope	Half-life, days[a]	Fractional production of parent, Z, atoms/fission	Fraction remaining after 50 000 hours, $\left(\frac{1 - e^{-\lambda t}}{\lambda t}\right)$	Fractional yield of parent after 50 000 hours, Y, atoms/fission	Daughter	Fractional yield of daughter after 50 000 hours, Z - Y, atoms/fission
			Uranium-233				
85	Kr	10.6 yr	$Z_2 = 3.7\times10^{-3}$	0.835	3.1×10^{-3}	Rb	6×10^{-4}
89	Sr	50	6.14×10^{-2}	.0346	2.1	Y	5.93×10^{-2}
91	Y	58	6.61	.0402	2.6	Zr	6.35
95	Zr	65	6.05	.0450	2.7	Nb(35 days)	5.78
95	Nb	35	5.78	.0242	1.4	Mo	5.64
103	Ru	40	1.30	.0277	4×10^{-4}	Rh	1.26
106	Ru	365	2.15×10^{-3}	.248	5.3	Pd	1.62×10^{-3}
125	Sb	985	2.05	.525	1.08×10^{-3}	Te	9.7×10^{-4}
127	Te	105	2.9	.0730	2×10^{-4}	I	2.7×10^{-3}
141	Ce	33	6.60×10^{-2}	.0229	1.5×10^{-3}	Pr	6.45×10^{-2}
144	Ce	285	4.19	.196	8.2	Nd	3.37
147	Pm	950	1.72	.514	8.9	Sm	8.3×10^{-3}
155	Eu	622	3.5×10^{-4}	.389	1.4×10^{-4}	Gd	2.1×10^{-4}
			Uranium-235				
85	Kr	10.6 yr	$Z_2 = 2.9\times10^{-3}$	0.835	2.4×10^{-3}	Rb	5×10^{-4}
89	Sr	50	4.50×10^{-2}	.0346	1.6	Y	4.34×10^{-2}
91	Y	58	5.60	.0402	2.3	Zr	5.37
95	Zr	65	6.78	.0450	3.1	Nb(35 days)	6.47
95	Nb	35	6.47	.0242	1.6	Mo	6.31
103	Ru	40	3.48	.0277	1.0	Rh	3.38
106	Ru	365	9.5×10^{-3}	.248	2.4	Pd	7.1×10^{-3}
125	Sb	985	2.1	.525	1.1	Te	1.0
127	Te	105	8.0	.0727	6×10^{-4}	I	7.4
141	Ce	33	6.10×10^{-2}	.0229	1.4×10^{-3}	Pr	5.96×10^{-2}
144	Ce	285	5.13	.196	1.00×10^{-2}	Nd	4.13
147	Pm	950	2.40	.514	1.23	Sm	1.17
155	Eu	622	5×10^{-4}	.389	2×10^{-4}	Gd	3×10^{-4}

[a]Except for Kr which has half-life in yrs.

The fractional yields of the radioactive parent isotopes and the daughter isotopes are given in table IV.

The fractional yields were calculated on the basis of a 50 000-hour reactor operation. However, for pressure calculation purposes they do not change appreciably for reactor operating times from 2000 hours (approx. 90 days) to 100 000 hours (approx. 11.5 years). For these operating times the xenon and cesium fractional yields are virtually the same and are the largest contributors to the pressure. Also, for krypton and rubidium, which are the other major contributors, there is only about a 1 percent change in the fractional yield between 2000 and 100 000 hours. Furthermore, an increase in the fractional yield of krypton will be accompanied by a decrease in rubidium, and vice versa, since radioactive krypton decays into rubidium. The pressure changes due to the changes in fractional yields of these two elements will thus tend to cancel out. Therefore, for pressure calculations, the fractional yields calculated in this report are valid for reactor operating times from 2000 to 100 000 hours.

Fission-Product Yields by Elements

For the pressure calculations, the individual isotopic yields were grouped and added together by elements. The complete list of fission-product yields by elements X is presented in table V.

All the elements were originally considered as potential contributors to the total pressure. Many of the elements in the list have comparatively low vapor pressures or yields, however, and do not make significant contributions to the total pressure. These were omitted from the final pressure calculations. The nine elements which do make significant pressure contributions and which were used in the final pressure calculations are listed in table VI.

TABLE V. - FISSION-PRODUCT YIELDS FROM FISSIONING OF URANIUM-233 AND URANIUM-235 INDUCED BY FISSION-SPECTRUM NEUTRONS (LISTED BY ELEMENTS)

Element	Mass numbers	U^{233}	U^{235}
		Fractional yield, X, atoms/fission	
Ge	72, 73, 74, 76	3.5×10^{-4}	1.2×10^{-4}
As	75	1.2×10^{-4}	5×10^{-5}
Se	77 to 80, 82	1.25×10^{-2}	0.64×10^{-2}
Br	81	.44	.24
Kr	83 to 86	5.46	4.02
Rb	85, 87	5.14	3.96
Sr	88, 89, 90	11.41	8.86
Y	89, 91	6.19	4.57
Zr	91 to 96	31.97	31.08
Mo	95, 97, 98, 100	20.44	24.91
Nb	95	.14	.16
Tc	99	4.75	6.13
Ru	101 to 104, 106	6.07	12.84
Rh	103	1.26	3.38
Pd	105 to 108, 110	.92	3.01
Ag	109	.15	.15
Cd	111 to 114, 116	.57	.20
In	115	.10	.03
Sn	117 to 122, 124, 126	1.07	.65
Sb	121, 123, 125	.34	.20
Te	125 to 128, 130	3.32	4.52
I	127, 129	1.84	2.64
Xe	131, 132, 134, 136	19.80	21.95
Cs	133, 135, 137	16.88	18.39
Ba	138	6.50	6.70
La	139	6.60	6.70
Ce	140, 141, 142, 144	13.97	13.34
Pr	141	6.45	5.96
Pm	147	.89	1.23
Nd	143 to 146, 148, 150	15.62	18.94
Sm	147, 149 to 152, 154	2.56	3.78
Eu	153, 155	.14	.21
Gd	155 to 158, 160	6×10^{-4}	7.4×10^{-4}
Tb	159	2×10^{-5}	3×10^{-5}
Dy	161	5×10^{-6}	5×10^{-6}
Total		1.924	2.095

TABLE VI. - SIGNIFICANT[a] FISSION-PRODUCT YIELDS FROM FISSIONING OF URANIUM-233 AND URANIUM-235 INDUCED BY FISSION-SPECTRUM NEUTRONS

Element	Mass numbers	U^{233}	U^{235}
		Fractional yield, X, atoms/fission	
Xe	131, 132, 134, 136	19.80×10^{-2}	21.95×10^{-2}
Kr	83 to 86	5.46	4.02
Cs	133, 135, 137	16.88	18.39
Rb	85, 87	5.14	3.96
I	127, 129	1.84	2.64
Te	125 to 128, 130	3.32	4.52
Br	81	.44	.24
Se	77 to 80, 82	1.25	.64
Cd	111 to 114, 116	.57	.20

[a]Omitted are fission products which have comparatively low vapor pressure or yield.

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